Effect of nanoglass grain size investigated by a mesoscale variable characteristic strain model

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ABSTRACT

Severe shear localization in metallic glasses (MGs) significantly limits their mechanical performance. Nanoglass (NG), composed of heterogeneous glassy domains created by introducing interfaces into MGs at the nanoscale, could be a promising strategy against severe shear localization, as demonstrated by numerous atomistic simulations. This study introduces a novel mesoscale kinetic Monte Carlo (kMC) model with a variable characteristic strain (VCS) to investigate the grain size effect in NGs. This model captures the complex evolution of shear bands during deformation, revealing a surprising transition from inhomogeneous to homogeneous deformation as the NG grain size decreases to approximately 10 nm. This transition is attributed to the impediment of shear band formation by the small grain size, facilitated by softer interfaces guiding early shear transformation zone (STZ) activities across the entire sample. Furthermore, a progressive reduction of elastic constants simulates the failure response observed in experiments. Our model predicts a critical grain size for the transition in agreement with experiments. This mesoscale model enables the investigation of NG deformation with microstructural features on an experimentally-relevant spatial-temporal scale. This paves the way for tailoring NG microstructures to achieve enhanced mechanical performance, and opens new avenues for exploring the influence of interfaces in controlling shear localization.

1. Introduction

The mechanical and physical properties of metallic glasses (MGs) are diverse due to their amorphous structure. Owing to the lack of long-range order in amorphous alloys, crystalline defects, such as dislocations and stacking faults, do not occur in MGs. As a result, MGs exhibit numerous favorable characteristics such as high yield strength, hardness, and fracture toughness, among others [1]. At room temperature, plastic deformation is highly localized in a few narrow planar defects known as shear bands [2,3]. Subsequently, the single shear band leads to catastrophic failure, which is a major obstacle to the application of MGs. Therefore, improving the ductility of MGs is a key issue of interest in materials science. The most intuitive route is to prevent the propagation of primary shear bands through amorphous alloys [4,5]. Experiments have reported that a single shear band can be hindered by multiple shear bands to further enhance the global plasticity of MGs [5-8].

A new amorphous alloy known as nanoglass (NG), synthesized by inert gas condensation, was first developed by Jing et al. [9]. Previous experiments have shown that MGs display a more pronounced shear band, while NGs exhibit excellent ductility with a few or multiple finer shear bands observed [10-13]. NGs comprising several glassy clusters have attracted considerable attention because of the enhancement of ductility [14-16]. Each dense glassy cluster in NGs, similar to a grain, is connected by loose glassy clusters, similar to interfaces. On the other hand, low density [17] and excess free volume [9,18-20] are characteristics of glassy interfaces in NGs, as first described by Sopu et al. [20].
The high free volume results in soft regions, which tends to reduce the resistance to deformation and serve as potential sites for shear transformation zones (STZs) [19,21–24]. Theoretically, the deformation at these sites could potentially form multiple shear bands rather than a single dominant shear band that typically observed in MGs. As a result, the existence of interfaces in NGs may delay the formation of a single shear band and improve their ductility [25,26].

Molecular dynamics (MD) simulations have revealed that the interfaces of NGs, rather than the grains, exhibit a greater capacity to accommodate plastic deformation [19,22,27]. In addition, early plastic deformation is preferentially distributed to the interfaces when NGs are subjected to tensile loading (28–36) or compressive loading [37]. MD simulations additionally revealed that grain size effects are significant in NGs [28–32,35,38,39]. The large grains, which are accompanied by a lower volume fraction of interfaces, cause a local softening behavior due to the formation of shear bands. In contrast, the finer grains associated with a higher volume fraction of interfaces cause more homogeneous deformation because the shear band has a finite thickness. Although MD simulations have provided relevant results on grain size effects, MD simulations still suffer from some inherent limitations, such as the time and length scales, which are typically limited to ~10 ns and 100 nm, respectively.

On the other hand, continuum simulations have revealed that the effects of the initial free volume distribution would affect the formation of shear bands in MGs [40]. In addition, the initial deformation in MG composites preferentially occurs in regions with a high free volume concentration [41]. A non-local plasticity model using finite element analysis revealed that plastic deformation in NGs can be retarded by a fine grain size [42]. These simulations emphasize that NG interfaces with excessive free volume can contribute significantly to the nucleation of shear bands. In fact, regions with different amounts of free volume can be treated essentially as a composite material problem [24,43,44]. However, these continuum methods, which are mainly based on the free volume theory, usually require the derivation of complex constitutive laws to describe the relationships between stress and strain during plastic deformation [45–47]. In addition, examining the microstructure evolution during deformation is difficult.

The kinetic Monte Carlo (kMC) mesoscale models [48–51], which often arise in the context of Eshelby’s inclusion problem [2,52], could be used to trace the evolution of plastic deformation from embryonic to mature shear bands in MGs. In addition, these mesoscale models are well suited to composite material problems in MGs, the second phase of metallic glass matrix composites can be characterized by the number of transformation modes and the magnitude of the Helmholtz free energy barrier for STZs. For example, one model has been extended to investigate the influence of medium-range order (MRO) in MGs [53,54]. In general, a characteristic STZ strain in the form of a constant on the order of ~0.1 [2,55] is used in STZ dynamics. However, the magnitude of the characteristic strain depends on the glass compositions and structural states [2,55]. Mesoscale simulations have found that the magnitude of the characteristic strain is responsible for different properties of the STZ in MGs [53]. Therefore, applying a constant STZ characteristic strain to MGs may not realistically simulate the structural evolution of MGs due to plastic deformation.

Researchers have focused their attention on investigating the plastic behavior in MGs through simulations for decades. However, there have been some unresolved issues observed in such simulations, such as the failure criterion in MGs. Hardin and Von Mises [36] utilized a mesoscale kMC model developed by Homer and Schub [49] to investigate MG matrix composites. The maximum shear strain was assigned as the failure criterion by Hardin et al. When the shear strain of any element reached the critical value, it was considered to be a system failure. However, the system failure caused by the accumulative shear strain could not reasonably explain the failure initiation. On the other hand, free-volume continuum simulations have defined other failure criteria, such as a critical concentration of free volume [57–59] or nanovoids [60]. Once the concentration of free volume or nanovoids in an element reaches the critical value, the element is treated as a local failure instead of a system failure. However, owing to the lack of constitutive laws, the mesoscale kMC model cannot utilize internal state variables such as free volume to account for the local failure.

In this study, a mesoscale kMC model where the magnitude of the characteristic strain of each STZ varies according to the state function is demonstrated. The magnitude of the characteristic strain is described by a sigmoid function based on the von Mises strain during the deformation process. The state function not only controls the variation of the characteristic strain, but also introduces a reduction of progressive stiffness at the STZs when the corresponding von Mises strain reaches a certain value. The proposed mesoscale kMC model is applied to investigate the evolution of deformation in NGs with different grain sizes and in bulk MGs.

The paper is structured as follows. Section 2 presents the modified activation energy barrier for the mesoscale kMC model based on the variable characteristic strain (VCS). The reduction of elastic constants of STZs is explained under the condition of VCS. In addition, the characterizations of interfaces and grains are revealed by different parameters. Section 3 provides a detailed description of numerical examples and simulation results, focusing on the effects of VCS on mechanical behaviors. Furthermore, the reduction of elastic constants of STZs under the condition of VCS is also investigated. Section 4 presents the deformation mechanism of grain size effects and the corresponding parametric studies. The studies include differences in the Helmholtz free energy barriers between interfaces and grains, the parameters associated with the proposed sigmoid state function, and the reduction factor of the elastic constants. Finally, conclusions on the grain size effects of NGs are summarized in Section 5.

2. Methodology

In this section, we describe the main aspects of our proposed material model. Section 2.1 introduces the VCS, which varies with the increasing von Mises strain in each STZ to characterize the shear band propagation states. The modified activation energy barrier is applied in the mesoscale kMC model. Section 2.2 displays the reduction factor of elastic constants proposed to model the local mechanical behaviors of STZs during deformation. In particular, the reduction of elastic constants of STZs is under the condition of VCS. Section 2.3 presents the kMC modeling framework for MG and NGs with different grain sizes. The NGs with different grain sizes are determined by the arrangement of voxels. Different transformation modes and Helmholtz free energy barriers are used to distinguish between interfaces and grains of NGs.

2.1. Variable characteristic strain

In the early stages of deformation, the stress state within the MG may not be sufficiently concentrated to nucleate and sustain a shear band. Nevertheless, with the progress of deformation, stress concentrations may develop around defects or microstructural features, leading to the nucleation and growth of a shear band. At this stage, the shear band may grow slowly as it accumulates deformation and strain softening within a localized region of the MG. With the growth of the shear band and further progress of deformation, the stress state within the MG can become increasingly localized and concentrated within the shear band, leading to the high localization of deformation within the band and resulting in rapid propagation and further strain softening. Additionally, with the propagation of the shear band, damage can be accumulated in the form of microcracks, voids, or other defects. This damage can reduce the strength and toughness of the shear band and lead to catastrophic failure once a critical level of damage is reached.

To characterize the stages of shear band propagation, Shimizu et al. [61] proposed the aged-rejuvenation-glue-liquid (ARGL) shear band model in which the shear band propagation is progressive. In the ARGL
shear band model, the shear band propagation can be developed into rejuvenation, alienated glass, and near liquid after the elastic deformation (i.e., well-aged state). In addition, the mesoscale kMC model of Homer revealed that the shear band evolution can also be divided into several stages [62]. The multiple stages make it easier to understand the formation of the shear band [63,64]. The exact behavior of shear bands may depend on the material and specific deformation conditions, and further research is required to completely understand this complex phenomenon.

The characteristic strain tensor, on the other hand, refers to the amount of shear deformation that a material can withstand with stable local shear deformation. Based on previous numerical simulations [65-67] showing that the evolution of the free volume concentration is similar to a sigmoid function, we believe that the magnitude of the characteristic strain tensor increases with the accumulation of different shear transformation modes in the process of MG deformation. We performed some MD simulations which were described in the supplementary data to verify this idea. Therefore, the evolution of the characteristic strain in this study is considered as a progressive process and follows a sigmoid function depending on the accumulation of deformation, where the deformation is described by the von Mises strain [48]. The total characteristic strain, denoted by the VCS, can be divided into two parts as follows:

\[ (ε_0)_y^{(m)} = (ε_0)_y^{(m)} + f(ε^M) (ε_0)_y^{(m)}, \]  
(1)

where the first term \((ε_0)_y^{(m)}\) is the intrinsic characteristic strain for the \(m\)-th mode, which is usually considered to be \(0.1\). The second term is the original characteristic strain multiplied by the sigmoid function \(f(ε^M)\), which varies with the local von Mises strain \(ε^M\), and is expressed in Eq. (2).

\[ f(ε^M) = \frac{f_{\text{max}}}{1 + \exp(-C(ε^M - 0.5))} \leq f_{\text{max}}, \]  
(2)

where \(f_{\text{max}}\) is the maximum value of the sigmoid function and is assumed to be 0.2. Parameter \(C\) is used to adjust the slope of the sigmoid function. The amplitude of VCS varies with the von Mises strain and can be divided into four stages (as shown in Fig. 1(a)).

Owing to the excellent performance of the Zhao’s kMC model in the formation of shear bands in MGs [48], the concept of the generation-dependent landscape in this model was adopted here to study the plastic deformation of MGs. As the strain tensor is proportional to the characteristic strain, and according to the VCS, the activation energy barrier can be modified as follows:

\[ Q^{(m)} = ΔF \exp\left(-η_s\right) - \frac{1}{2} V_p σ_y (1 + f(ε^M) \exp\left(-η_s\right)) (ε_0)_y^{(m)}, \]  
(3a)

\[ η_s = η_s \exp\left(-\frac{t_{\text{elap}}}{τ}\right), \]  
(3b)

where \(Q^{(m)}\) is the activation energy barrier, \(ΔF\) represents the Helmholtz free energy barrier, \(V_p, σ_y\), and \(ε_0\) denote the volume, stress tensor, and characteristic strain tensor, respectively. In addition, \(η_s\) is the generation-dependent relaxation recovery parameter and \(η_s\) is a user-defined parameter which controls the softening behaviors; \(t_{\text{elap}}\) is the time elapsed since the last event was triggered at the same location. Increasing \(η_s\) increases the relaxation recovery parameter, \(η_s\), and thus increases the activation energy barrier. Our numerical experience suggests that the value of \(η_s\) can be set to 1.0 (More details are provided in the supplementary data.).

2.2. Reduction in the elastic constants of STZs

The elastic constants of MGs under plastic deformation can have the generation-dependent reduction, thus the elastic constants decrease with the increase in strain. Experimental studies using nanoindentation [68,69] and quantitative nanomechanical measurements (QNM) [70, 71] have revealed that the Young’s modulus of MGs decreases significantly near shear bands. This reduction in Young’s modulus is attributed to changes in the local atomic structure and bonding caused by the shear band deformation.

In this study, the reduction in the elastic constants for STZs was adopted to characterize the evolution of bearing failure. For isotropic materials, only two elastic constants are independent and the effects of Poisson’s ratio on the mechanical behaviors are not obvious from our numerical experiences. Therefore, we assume that Young’s modulus and shear modulus decreased with the same amplitude during the deformation while Poisson’s ratio remained constant. The reduction in the elastic constants is assumed to be progressive and in the form of a sigmoid function. In addition, the reduction is assumed to be initiated after the STZ has progressed through the full four stages of VCS shown in Fig. 1(a). The evolution of Young’s modulus is expressed in Eq. (4) and shown in Fig. 1(b). The associated reduction is discussed in Section 4.4.

\[ EC(ε^M) = \left(1 - \frac{R_{\text{EC}}}{1 + \exp(-C(ε^M - 1.5))}\right) \exp\left(-η_s\right) \]  
EC,  
(4)

where \(EC\) are the modified elastic constants, i.e., Young’s modulus and shear modulus, enacted after the von Mises strain is greater than 1.0, \(EC\) are the original elastic constants, and \(R_{\text{EC}}\) is the reduction factor of the elastic constants. It is worth noting that the generation-dependent relaxation recovery is taken into account in the reduction of the elastic constants.

![Fig. 1](image-url)

Fig. 1. (a) Variation of the characteristic strain amplitude (corresponding to \(f_{\text{max}} = 0.2\)) with the von Mises strain while maintaining a constant amplitude of the characteristic strain during the deformation. According to the von Mises strain, the sigmoid function can be divided into four stages. (b) The reduction of elastic constants is assumed to follow a sigmoid function. When the von Mises strain of STZs is greater than 1.0, the amplitude of the elastic constants begins to decrease. The reduction in the elastic constants of STZs would increase with increasing von Mises strain of STZs until the amplitude of the elastic constants reaches 1. \(R_{\text{EC}}\).
2.3. Numerical models

To verify the accuracy of the proposed computational method, tensile test simulations were performed using the strain control method on two-dimensional square specimens composed of zirconium-based MGs and NGs at a temperature of 300 K [72]. The strain rate and strain increment are $1 \times 10^{-4}$ s$^{-1}$ and $1 \times 10^{-4}$, respectively. The length of these square specimens is 240 nm, and the samples are discretized into $240 \times 240$ voxels. Periodic boundary conditions are applied to these four edges. The dimensions of the STZs are assumed to be $1 \times 1 \times 1$ nm, and the STZs are divided into interfaces and NG grains according to their physical properties. MGs and NGs with different grain sizes were considered.

The parameters for grains and interfaces are listed in Table 1. To simplify the calculation of the stress field, an identical Young's modulus and Poisson's ratio are initially assigned to interfaces and grains. Notably, in the mesoscale model, MGs can be considered as a single phase without any interfaces. The Helmholtz free energy barriers for grains $\Delta F_g$ and interfaces $\Delta F_i$ are $3.5$ eV and $1.5$ eV, respectively, for each STZ [72]. In order to characterize the heterogeneities in MGs and NGs, the heterogeneously randomized transformation modes [48] are introduced in this study. Since the softer interfaces are constrained by the harder grains, it is assumed that there are fewer transformation modes for interfaces ($m = 5$) than for grains ($m = 40$), schematically shown in Fig. 2(a). To investigate the effect of grain size on NGs, the grain sizes of the hexagonal grains [31,33,42,73,74] are assigned as 6 nm, 10 nm, 18 nm, and 38 nm, and the volume fractions of the interfaces are 43.75 %, 30.56 %, 19.00 %, and 9.75 %, respectively. A typical single grain of 6 nm composed of STZs is shown in Fig. 2(b). The width of the interface, which is independent of the grain size, in the vertical direction consists of two STZs, i.e., 2 nm [75,76]. The NG samples with different grain sizes are shown in Fig. 2(c).

### Table 1

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<tr>
<th>Parameters</th>
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<th>Grain</th>
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<td>Poisson's ratio [72]</td>
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<td>Helmholtz free energy barriers, $\Delta F$</td>
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3. Results

In this section, the mechanical behaviors of MG and NGs with different grain sizes are measured by the stress-strain (SS) curves and von Mises strain distributions in tensile tests. In Section 3.1, the simulation results with and without the VCS are compared to investigate the effect of VCS on the SS curves and the evolution of von Mises strain distributions. Section 3.2 introduces the strain localization parameter [77] to interpret the influence of VCS on the grain size effects of MG and NGs at different deformation stages in the mesoscale models. Section 3.3 discusses the reduction in elastic constants of STZs to model the bearing failure in tensile tests of MG and NGs under the condition of VCS.

3.1. Variable characteristic strain effects

Fig. 3(a) and (b) show the SS curves of MG and NG specimens under uniaxial tensile testing without and with consideration of VCS, respectively. All the SS curves in Fig. 3 exhibit an identical slope during elastic deformation because of the identical Young’s moduli used for MG and NGs. The apparent yield stress, defined by the maximum stress without any deviation from the linear region, is 3.71 GPa for the MG. The apparent yield stresses of NGs are 1.54 GPa, 1.43 GPa, 1.35 GPa, and 1.30 GPa for the grain sizes of 38 nm, 18 nm, 10 nm, and 6 nm, respectively. Clearly, the apparent yield stresses of MGs are considerably greater than those of NGs, and the finer the grain size of the NGs, the lower the apparent yield stress. VCS has a negligible influence on the apparent yield stress.

For the cases without VCS, the peak strengths of NGs are lower than that of MG and decrease with decreasing grain size. One can observe that the MG quickly exhibits typical strain softening behavior, whereas NGs exhibit superplastic-like behaviors. Previous atomistic simulations in the literature have revealed that NGs can undergo pronounced strain hardening or superplastic-like deformation when the grain size is sufficiently small [28,32]. On the other hand, NGs with larger grain sizes exhibit a greater propensity for softening in comparison to smaller grain sizes. However, the effect of grain size on the trends of SS curves is less pronounced for NGs in the absence of VCS.

When VCS is considered, the SS curves display greater softening after reaching the apparent yield stress compared to simulations without VCS. When comparing MG with and without VCS, there is a significant difference in the magnitude of the stress drop. It was found that as grain size decreases between 38 nm and 10 nm in NGs, the magnitude of the stress drop also decreases. Meanwhile, the SS curve of NG with a grain size of 6 nm exhibits superplastic-like behaviors. In other words, NGs exhibit increasing softening characteristics as the grain size increases from 6 to 38 nm. These features of the SS curves under VCS are consistent with the results reported in previous studies that used MD simulations with periodic boundaries [31,35]. In MD simulations, the presence of the free surface results in a more pronounced stress decrease than in models without free surfaces [28,29,32].

The local von Mises strain distributions during the strain-controlled deformation in each MG and NG sample corresponding to the cases without VCS and with VCS are shown in Figs. 4 and 5, respectively. Without considering VCS, STZs in the MG sample are distributed randomly at different locations as early-stage nuclei of immature shear bands, as seen in Fig. 4 [3,78]. As the applied strain increases, the percolation of STZs in the MG sample penetrates through the model to form shear bands. In contrast to MG, the NGs exhibit softer structural domains due to the interfaces between NG grains; hence, the interfaces are preferential sites for the local plastic deformation. These results are consistent with MD simulations reported previously [28–32]. When the level of plasticity in NGs exceeds the capacity of the interfaces, the percolation of STZs extends into the NG grains, resulting in more widely distributed active shear zones involving STZ activities. The different results between MGs and NGs demonstrate the advantages of NGs in resisting shear localization by using interfaces to disperse STZs. The NG with 6 nm grain size is superior in inducing homogeneous deformation. The interfaces of NGs succeed in being potential sites for STZs. However, when the applied strain reaches 0.2, there is no clear transition from inhomogeneous to homogeneous deformation with decreasing grain size, as shown in Fig. 4. In the cases without VCS, the softening behavior of NGs exhibits a weak dependence on grain size, as shown in Figs. 3(a) and 4.

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During the early stage (i.e., applied strain = 0.04–0.08), it can be observed that the STZs in the MG sample in Fig. 5 have an almost identical pattern to the MG sample without VCS in Fig. 4. Similarly, as the applied strain increases, an accumulation of plastic deformation results in the formation of a shear band. Notably, the shear band in the MG with VCS is more concentrated than that without VCS. The impact of VCS on the SS curves and the evolution of von Mises strain distributions in tensile tests. In Section 3.1, the simulation results with and without the VCS are compared to investigate the effect of VCS on the SS curves and the evolution of von Mises strain distributions. Section 3.2 introduces the strain localization parameter [77] to interpret the influence of VCS on the grain size effects of MG and NGs at different deformation stages in the mesoscale models. Section 3.3 discusses the reduction in elastic constants of STZs to model the bearing failure in tensile tests of MG and NGs under the condition of VCS.
the NG (18 nm) with VCS, a shear band is eventually formed, which runs across the NG grains at this severe deformation stage, but its shape is not as complete as the band seen in the MG and NG (38 nm) samples. Although the interfaces cannot halt the initiation of the shear band, they can retard and disturb its development. Hence, if the grain size is sufficiently small, the shear band formation is almost suppressed, as seen in NGs (6 nm). The correlation of the shear band development with the grain size effect corresponds to the softening to superplastic-like transition observed in the SS curves. This transition is successfully captured in the NGs with VCS, but not without considering VCS.

3.2. Strain localization parameter

To further verify the different mechanisms at play in NGs and MGs, a strain localization parameter (SLP) [77] was used to analyze the local plastic strain accumulation during the applied tensile strain process. The SLP is defined as follows:

$$\phi(\varepsilon_i^M) = \frac{\sum_{i=1}^{N} (\varepsilon_i^M - \varepsilon_{\text{avg}}^M)^2}{N},$$

where $N$ is the total number of voxels, $\varepsilon_i^M$ is the von Mises strain of the $i$-th voxel, and $\varepsilon_{\text{avg}}^M$ is the average of the von Mises strain of all voxels. A higher value of the parameter $\phi$ indicates more localized deformation and vice versa. The SLP evolution for MGs and NGs with grain sizes of 38, 18, 10, and 6 nm are displayed in Fig. 6, (a) without VCS and (b) with VCS. These figures are divided into three deformation zones: applied strains of 0.00–0.05, 0.05–0.10, and 0.10–0.20.

The SLP curves without VCS for MG and NGs are shown in Fig. 6 (a). In zone I, the increase in the SLP of NGs is similar for the four grain sizes, and starts earlier than in MG. These phenomena indicate that, compared to the grain areas, the interfacial areas indeed induce an earlier initiation of STZ, and that STZs are distributed along the predetermined geometric framework. Although the SLP of MG starts later, it is more significant than the SLP of NGs. The SLP curve of MG rapidly grows with increasing strain.
beyond the curves of NGs in zone II, confirming that NG is an effective strategy in suppressing the development of shear localization. However, only a minor difference exists among the four SLP curves at the end of zone III following the order of 38 nm > 18 nm > 10 nm > 6 nm.

Fig. 6 (b) shows the SLP curves for MG and NGs with different grain sizes considering VCS. VCS does not change the primary mechanisms of MG and NGs in zone I, but does create distinct grain size effects when accessing zone II and zone III. Secondary growth of the slope of the MG SLP curve is observed in zone II. In addition, the slope of the MG SLP curve is steeper with VCS than without it. Similar to Fig. 6 (a), the SLP curve of MG rapidly grows beyond the SLP curves of NGs in zone II. On the other hand, the SLP curves of NGs at all grain sizes exhibit only marginal differences in zone II. However, compared with that of the 18 nm, 10 nm, and 6 nm NGs, the SLP curve of NG (38 nm) exhibits a sudden upward deviation in zone III. This upward turning accounts for the transition of the STZ development in Fig. 5, in which the capacity of the interfacial network in NG (38 nm) is not sufficient to prevent the eventual development of the shear band. According to Fig. 6 (b), 10 nm is an effective grain size for suppressing shear localization.

3.3. Reduction in elastic constants

In this section, the reduction in the elastic constants of STZs according to Eq. (4) is introduced to account for the bearing failure behavior of MG and NGs with VCS in tensile tests. The $R_{EC}$ is set to 5%, which represents the upper bound of the reduction in the elastic constants, and is substituted into Eq. (4). The effect of the reduction value is addressed in Section 4.4. The SS curves with reduced elastic constants (using $R_{EC}$ in Eq. (4)) for MGs and NGs are shown in Fig. 7. The SS curve with the reduction in elastic constants exhibits an extremely sharp drop into the plastic region after passing the point of maximum strength, closer to the fracture behavior observed in experiments. Notably, the extremely sharp drop in the SS curve of MG obtained by considering the reduction in elastic constants indicates that the MG has lost its load-bearing capacity. Although it is possible to continue the tensile tests in numerical simulations, it lacks any physical significance, so we have terminated the simulations by marking it with an ‘X’ on the SS curve.

Such a bearing failure feature is also observed in the SS curves with the reduction in elastic constants for NGs, but it manifests different mechanical responses to grain size during the failure regime.
Fig. 5. Distributions of the von Mises strain with VCS for MG and NGs with grain sizes of 38, 18, 10, and 6 nm under applied strains of 0.04, 0.06, 0.08, 0.10, 0.15, and 0.20. The white regions represent the absence of plastic deformation. The shear band in the MG with VCS is more concentrated than that without VCS. For NGs, the shear band formation is almost suppressed if the grain size is sufficiently small. Distributions of the von Mises strain with VCS show a transition from inhomogeneous to homogeneous deformation.

Fig. 6. Strain localization parameter curves for MG and NGs with grain sizes of 38, 18, 10, and 6 nm are depicted for two cases: (a) without VCS and (b) with VCS. The figures are divided into three deformation zones corresponding to applied strains of 0.00–0.05, 0.05–0.10, and 0.10–0.20. A higher value of the strain localization parameter indicates more localized deformation and vice versa.
example, the plastic regime in the SS curve of NGs (38 nm) is considerably longer than that of MGs, but shorter than that of NGs (18 nm, 10 nm, and 6 nm). In addition, the slope of the SS curve in the failure regime changes from steep to gentle with the decrease in grain size from 38 to 6 nm, indicating a brittle-to-ductile transition. Clearly, the elastic constants reduction factor in the model can accentuate the failure features on the SS curve of not only MGs but also NGs with a size-dependent effect. Notably, the mesoscale model used here is still in the realm of continuum models such as the finite-element method; hence, a practical fracture as seen in the studies of MD cannot be shown in the model.

The illustration in Fig. 7 depicts SS curves of MG and NG specimens in experimental tensile tests [79]. Based on the experimental results of Wang et al. [21] and Yang et al. [79], it is evident that NG specimens exhibit increased plasticity compared to MG specimens in the SS curves, attributed to the presence of multiple shear bands [21]. In addition, NG samples exhibit necking behavior under tensile loading, reducing cross-sectional area until failure [21,79]. In contrast to NG, MG specimens fail in a brittle manner as expected. In our numerical model, the SS curves of MG and NGs, shown in Fig. 7, demonstrate a transition from brittle to ductile behavior due to the reduction in elastic modulus, in agreement with the observed experimental results [21,79].

4. Discussion

In this section, the in-depth insight of MG and NGs with different grain sizes in tensile tests is discussed. In addition, related parametric studies are carried out to explore how the parameters in our proposed model affect the mechanical behaviors of MG and NGs. Section 4.1 reveals the deformation mechanisms of MG and NGs based on the proposed models. It explains the relationship between the thickness of shear bands and the grain sizes of NGs. A comparison between our results and experimental observations is made. Section 4.2 discusses the role of Helmholtz free energy barriers for grains ($\Delta F_g$) and interfaces ($\Delta F_i$) in the mechanical behaviors of NGs. The effect of the difference in energy barriers on the selection of plastic events in kMC is investigated. Section 4.3 presents parametric studies on $f_{max}$ and $C$ in the sigmoid function, showing how these parameters influence the SS curves. Section 4.4 presents parametric studies of the maximum reduction in elastic constants $R_{EC}$, which clarifies the reason for setting $R_{EC}$ to 5% in Section 3.3.

4.1. Deformation mechanism

According to the literature [2,80], homogeneous deformation in metallic glasses can be regarded as the viscous flow of a supercooled fluid, akin to what is typically observed in the supercooled liquid regime. The formation of shear bands results from the instability of the homogeneous deformation, which is primarily dominated by the free-volume-initiated process. Our study indicates that this homogeneous deformation instability scales with grain size and can be constrained by the NG interfaces. In particular, our simulation results demonstrate that the softer interfaces indeed play an important role in guiding early STZ activities to spread along the interfaces across the entire sample, thereby blunting and slowing down the typical shear-localization process in the initial stages. When the STZ activities extend beyond the interfaces and into the grains, the mechanisms may involve competition between the grain size and shear band incubation size, or interaction between the immature shear band and interfaces. The thickness of shear bands is typically narrow, around 10 nm [81].

This implies that as the grain size approaches this range, it is able to constrain the development of a shear band. Consequently, the nucleation of immature shear bands becomes possible, but these nuclei struggle to evolve into mature shear bands. Furthermore, the interaction between shear bands and interfaces becomes more effective, and the interfaces can effectively block the shear band propagation. To gain insight into the mechanism of these phenomena, we investigated the spatial correlation of von Mises strain at different applied strain levels after yielding. The spatial correlation is given by the following equation:

$$\rho(r) = \frac{1}{V} \sum_{i=1}^{N} e^{\epsilon(r_0)} e^{\epsilon(r_0 + r)}$$

where $e^{\epsilon(r_0)}$ is the von Mises strain of a reference position $r_0$, $e^{\epsilon(r_0 + r)}$ is the von Mises strain of a point at a distance $r$ from the reference position. $V$ is the total number of STZs. The spatial correlation curves of the von Mises strain at applied strains of 0.10, 0.15 and 0.20 are shown in Fig. 8. It can be observed that for the grain size of 38 nm NG, the spatial correlation decreases significantly with increasing distance. These results imply that the deformation in the grain size of 38 nm NG is quite non-uniform, thus mature shear bands have been developed. On the other hand, for the grain sizes of 6 and 10 nm, there are no significant changes in the spatial correlation with increasing distance. These results indicate that the deformation is relatively uniform at grain sizes of 6 and 10 nm, thus no mature shear bands exist. Our mesoscale model, applied at an experimental strain rate, aligns with findings in experiments [21]. Notably, when the grain size approaches the shear band thickness, NGs with finer grain sizes effectively impede shear band propagation, as the size of flow defects in NG is limited by grain size [21].

4.2. Activation energy barrier spectra

The initial Helmholtz free energy barriers of the interfaces and grains are assigned as 1.5 eV and 3.5 eV, respectively. The plastic behavior based on kMC theory would be influenced by the activation energy barrier spectra with the increase in applied strain. NGs with a grain size of 18 nm were selected as the benchmark. The evolution of the activation energy barrier spectra during the plastic deformation with VCS is shown in Fig. 9(a).

Under an applied strain of 0.005, the activation energy barrier spectra of the interfaces and grains are separated. The total activation energy barriers of the interfaces are considerably less than those of the grains. As the lowest activation energy barrier occurs at the interfaces, the plastic behavior of NGs is dominated by the interfaces, which could accommodate most of the plastic deformation to retard the formation of the primary shear band. With the increase in applied strain, the activation energy barrier spectra of both interfaces and grains are
broadened in the horizontal direction. Under an applied strain of 0.015, the activation energy barrier spectra of the interfaces and grains begin to overlap. However, most of the activation energy barrier spectra of the interfaces are still lower than those of the grains, indicating that the plastic behavior is still largely dominated by the interfaces. In kMC theory, the voxels with the lower activation energy barriers, in this case the interfaces, are the sites with the greatest potential for transformation.

The difference in the activation energy barriers between the interfaces and grains would directly affect the von Mises strain.

**Fig. 8.** The spatial correlation of von Mises strain for NGs at applied strains of 0.10, 0.15, and 0.20. For the grain size of 38 nm NG, the correlation coefficient decreases significantly with increasing distance. For NG (6 nm) and NG (10 nm), there are no significant changes in the spatial correlation with increasing distance.

**Fig. 9.** (a) Evolution of the activation energy barrier spectra of interfaces and grains for an NG grain size of 18 nm with applied strain values of 0.005, 0.010, 0.015, 0.030, and 0.045. To demonstrate the difference in the Helmholtz free energy barriers between the interfaces and grains, the $\Delta F_g$ is set to 3.5 eV, and the $\Delta F_i$ is set to 3.5, 2.5, and 1.5 eV in Fig. 9(b), Fig. 9(c), and Fig. 9(d), respectively. The corresponding von Mises strain distributions for NGs (18 nm) with VCS are shown in (b), (c), and (d) under an applied strain of 0.05. The gray lines represent the interfaces and the white regions represent the absence of plastic deformation.
distributions during plastic deformation. To understand how the difference in the Helmholtz free energy barriers between the interfaces and grains affects plastic deformation, the $\Delta F_{i}$ is set to 3.5 eV, and the $\Delta F_{i}$ of 3.5, 2.5, and 1.5 eV were investigated. Under an applied strain of 0.05, the corresponding distributions of von Mises strain for NGs (18 nm) with VCS under different combinations of $\Delta F_i$ and $\Delta F_i$ are shown in Fig. 9(b)–(d). In Fig. 9(b), $\Delta F_{i}$ and $\Delta F_{i}$ are 3.5 eV, and most of the plastic deformation is randomly distributed in the grains. On the other hand, in Fig. 9(c), when $\Delta F_{i}$ is reduced to 2.5 eV, more plastic deformation is clearly located at the interfaces, while some plastic deformation still occurs within the grains. When $\Delta F_{i}$ is reduced to 1.5 eV (Fig. 9(d)), most of the plastic deformation is accommodated by the interfaces. These results indicate that the difference in the Helmholtz free energy barriers between the interfaces and grains must be sufficiently large to make the interfaces to be the preferred sites for STZs in early deformation.

### 4.3. Effect of the sigmoid function on SS curves

In Eq. (2), a sigmoid function is used to describe the growth of the VCS, where the parameters $f_{\text{max}}$ and $C$ control the maximum value and slope of the sigmoid function, respectively. The effects of these two parameters on the SS curve are explored below using NGs with a grain size of 18 nm. Notably, in these simulations, the VCS is considered but without the reduction in Young’s modulus. The parameters $f_{\text{max}}$ and $C$ are taken to be 0.2 and 0.3, and 10 and 20, respectively. The sigmoid functions with different combinations of parameters $f_{\text{max}}$ and $C$ are shown in Fig. 10(a). As can be seen from Fig. 10(a), $f_{\text{max}}$ represents the magnification factor of the intrinsic characteristic strain, $\gamma_0$, in the VCS. For example, $f_{\text{max}} = 0.3$ indicates that the maximum value of VCS is 1.3 times the intrinsic characteristic strain. In addition, for higher $C$, when the von Mises strain is less than 0.25, the VCS increases slowly (stage 1 in Fig. 1(a)), but when the von Mises strain is between 0.25 and 0.75, the VCS increases dramatically (stages 2 and 3 in Fig. 1(a)).

The corresponding SS curves are shown in Fig. 10(b). From Fig. 10(b), the softening of the SS curves rendered by a smaller $C$ value is less obvious than those rendered by a higher $C$ value. In addition, the softening of the SS curves rendered by a higher $f_{\text{max}}$ value is faster than those rendered by a smaller $f_{\text{max}}$ value. As the propagation of the shear band is determined by the stored elastic energy [82], a higher $f_{\text{max}}$ will generate a higher VCS, resulting in the release of more elastic energy at the intrinsic shear band and a more localized shear band, making softening more obvious. On the other hand, a higher C value will impede the shear band propagation as the VCS value will be lower in the first stage. Therefore, less stored elastic energy will be released in the intrinsic shear band. Although the VCS produced by the higher C value increases rapidly in stages 2 and 3, the higher VCS is limited to a few regions. As a result, softening of the SS curves is not obvious.

### 4.4. Effect of the reduction factor

To examine the effects of elastic constants reduction, $R_{\text{EC}}$ in Eq. (4), on the SS curves of NGs, NGs with a grain size of 18 nm are taken as an example. Fig. 11(a) shows the SS curves for the cases of a 5, 10, and 20 % reduction in elastic constants. As can be seen from Fig. 11(a), the SS curves corresponding to three different reductions in elastic constants all coincide prior to entering the stress drop regime. Notably, the earliest stress drop occurs at 10 % reduction in Young’s modulus, while the largest stress drop occurs at 20 % reduction. The stress drop amplitudes do not closely follow the value of the reduced elastic constants. On the other hand, the applied strain at which significant stress drop occurs can represent the ability to accommodate plastic deformation. However, the applied strain at which the significant stress drop occurs does not correlate with the reduction factor.

To obtain more information on how the mechanical behaviors are affected by the elastic constants reduction, five numerical simulations were performed with different random seeds in the kMC model for a reduction factor of 5 % (Fig. 11(b)). From Fig. 11(b), before the significant stress drops occur, a marginal difference in the SS curves can be observed. Subsequently, significant stress drops with different random seeds occur at different applied strains, which is due to the stochastic nature of the kMC algorithm. This observation could explain the irregularity of the significant stress drops in Fig. 11(a). In other words, the stochastic nature of the kMC algorithm exerts a greater influence than the reduction factor of the elastic constants. Therefore, in Fig. 11(a), the significant stress drop occurs irregularly at different applied strains, and does not appear to be sensitive to the reduction factors. In addition, the reduction factor of elastic constants introduced to characterize the bearing failure ability is defined as 5 %. The SS curves in Fig. 7 show that a 5 % reduction in elastic constants is sufficient to simulate the bearing failure behavior.

### 5. Conclusions

Mesoscale kMC models have proven to be a powerful approach for studying the deformation behavior of metallic glasses by treating the shear transformation zone (STZ) as a basic deformation unit. In this study, we have delved into the grain-size effect on the mechanical response to uniaxial tensile testing of NGs using a mesoscale kMC model, where the grain size ranges from 38 nm to 6 nm. The mesoscale kMC model, originally proposed by Zhao et al. [48], integrates two important features: heterogeneously randomized STZ transformation catalogs and generation-dependent softening in the kMC algorithm.

Based on their model, we further introduce: (a) the incorporation of a variable characteristic strain (VCS) in the activation energy barrier for the STZ mode, aligning with the aged-rejuvenation-glue-liquid model; and (b) a reduction in the elastic constants of STZs to mimic failure behavior akin to the experimental tests.

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**Fig. 10.** (a) Sigmoid function with different combinations of parameters $f_{\text{max}}$ and $C$, which are 0.2 and 0.3; 10 and 20, respectively. (b) The corresponding SS curves with VCS for NGs with a grain size of 18 nm. High values of $f_{\text{max}}$ and $C$ increase the softening of the SS curves.
Simulations of both models, with and without the VCS feature, reveal that designing NGs with hard grains and softer interfaces between grains effectively suppresses the development of primary shear bands. While these results align with findings from molecular dynamics (MD) simulations and experiments, our study marks the first verification using mesoscale kMC models. The inclusion of VCS, in particular, allows for a more detailed representation of shear localization in NGs with different grain sizes compared to models without VCS.

The VCS stands out in highlighting distinct amplitude variations in strain localization during different deformation stages. Consequently, our modified model illustrates a smooth transition from inhomogeneous deformation with a shaped primary shear band in MG and coarser grain NGs to homogeneous deformation with more widely distributed active shear zones involving STZ activities when the grain size is within 10 nm — a transition consistent with previous MD simulations.

A nuanced understanding of this deformation transition mechanism reveals that softer interfaces play a crucial role in guiding early STZ activities along interfaces, blunting and slowing down the typical shear localization process in the early stages. As the grain size decreases to 10 nm — a typical size of the shear band thickness theoretically, the interaction between shear bands and interfaces becomes more effective, with the interfaces effectively blocking shear band propagation.

In summary, our mesoscale kMC model offers a comprehensive exploration of the deformation behaviors of NGs. The detailed curve of the VCS, easily validated through MD simulations, provides insight into the atomic inelastic strain distribution under corresponding structural states. A similar method can, in principle, be applied to the details of the reduction of the elastic constants of STZs, contributing to a more thorough understanding of these materials.

CRediT authorship contribution statement

Chih-Jen Yeh: Writing – original draft, Software, Formal analysis, Data curation. Chang-Wei Huang: Writing – original draft, Project administration, Methodology, Conceptualization. Yu-Chieh Lo: Writing – original draft, Project administration, Methodology, Conceptualization. Shigenobu Ogata: Writing – review & editing, Methodology, Conceptualization. Ding Yuan Li: Writing – review & editing, Supervision. Jason Shian-Ching Jang: Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ijmecsci.2024.108981.

Appendix A

In the following, the transformation modes of STZs, the softening mechanisms in mesoscale kMC models of MGs, and the kMC algorithm are briefly...
revealed.

A.1. Transformation modes

The number of transformation modes in mesoscale kMC models depends on the material system considered. While for crystalline materials, the number of transformation modes should theoretically be related to specific directions [83–86]; for amorphous materials, the number of transformation modes should be infinite to represent their diverse microstructure [49]. A simple mesoscale kMC model of MGs with six transformation modes was first developed by Bulatov and Argon [51]. To better represent the infinite deformation directions and accurately capture the deformation mechanism of MGs, Homer and Schuh [49] constructed a mesoscale kMC model using a continuous integral form. In a different approach, Zhao et al. [48] developed a mesoscale kMC model with heterogeneously randomized transformation modes. The model effectively demonstrates that finite transformation modes can characterize the diverse deformation directions in MGs. In addition, the transformation modes of MGs are defined as generation-dependent, as schematically shown in Fig. A1, which is related to the softening mechanism in the model used by Zhao et al. [48]. In the next section, the softening mechanisms of MGs in the mesoscale kMC model are presented.

Fig. A1. The schematic diagram illustrates the generation-dependent transformation modes of STZ in MGs. The dashed parallelograms represent different possible deformation events according to the activation energy barrier. Bulatov and Argon first developed a simple mesoscale kMC model [51], in which the activation energy barrier for each transformation mode can be expressed as follows:

\[
Q^{(m)} = \Delta F - \frac{1}{2} V \sigma_{ij} \epsilon_{ij}^{(m)},
\]

where \(\Delta F\) represents the Helmholtz free energy barrier, \(V\), \(\sigma\), and \(\epsilon\) denote the volume, stress tensor, and characteristic strain tensor that has different shear transformation modes for each STZ (as illustrated in Fig. A1) [48], respectively. The superscript \(m\) represents the \(m\)-th transformation mode [48]. The number of transformation modes can account for the structural diversity of amorphous alloys. However, a single and fixed activation energy barrier is not sufficient to characterize the softening behavior of MGs [50]. To address the above limitation of Eq. (A1), Li et al. [50, 66] extended the mesoscale kMC model of Homer and Schuh [49] by incorporating free volume theory into the Helmholtz free energy barrier. Similarly, Zhao et al. [48] proposed a generation-dependent softening mechanism in the evolutionary free energy barrier [48]. In Zhao’s model [48], the change in the Helmholtz free energy barrier from generation \(g\) to generation \(g + 1\) for each step can be denoted by \(\Delta F_{g \rightarrow g+1}\) and can be expressed as follows:

\[
\Delta F_{g \rightarrow g+1} = \Delta F \exp (-\eta_g),
\]

where \(\eta_g\) represents the softening parameter. In addition, the softening parameter that is used to describe the softening behavior and the partial recovery process of the corresponding element can be further divided into two terms as follows:

\[
\eta_g = \eta_p + \eta_f \exp \left(-\frac{t_{\text{elap}}}{\tau}\right),
\]

where \(\eta_p\) and \(\eta_f\) are the permanent softening and temporary softening parameters, respectively; \(t_{\text{elap}}\) is the time elapsed since the last event was triggered at the same location. In Eq. (A3), the characteristic relaxation time \(\tau\) for each diffusional relaxation process is expressed as follows:

\[
\tau = \frac{1}{\nu_0 \exp (\frac{-Q^{\text{act}}}{k_B T})}
\]

where \(\nu_0\) is the trial frequency, \(k_B\) is the Boltzmann constant, and \(Q^{\text{act}}\) is the energy barrier to trigger a diffusional relaxation process [88]. Finally, the activation energy barrier for each transformation mode can be expressed as follows:

\[
Q^{(m)} = \Delta F \exp (-\eta_g) - \frac{1}{2} V \sigma_{ij} \epsilon_{ij}^{(m)}.
\]
In this study, the maximum value of $\eta_g$ is set to $-\ln(0.85)$ to control the upper limit of the softening behavior.

### A.3. kMC algorithm

In the mesoscale kMC model, the evolution of deformation can be categorized as pure elasticity, plasticity, or athermal plasticity. When the activation energy barriers are all positive, the triggered event could be pure elasticity or plasticity, depending on the time increment for each step. The activation rates are obtained by substituting the modified activation energy barrier into the following equation:

$$k_i^{(m)} = \nu_0 \exp\left(-\frac{Q_i^{(m)}}{k_BT}\right).$$  \hfill (A6)

The time increment for each step is calculated by the following equation:

$$t = \frac{1}{\sum k_i^{(m)}}.$$  \hfill (A7)

Plastic events are triggered when the time increment is less than the maximum residence time (strain increment/strain rate) for each step. Then, the stress solver calculates a new stress field. A purely elastic event is triggered instead when the time increment is greater than the maximum residence time, indicating that only strain deformation is applied during this step.

### Appendix B

Our proposed model is developed on the basis of Zhao’s model [48]. If the variable characteristic strain (VCS) and the generation-dependent relaxation recovery parameter are ignored, our model degenerates to the same as the original mesoscale kMC model. Therefore, the original mesoscale kMC model can be used as a calibration model.

A square MG sheet with dimensions of 217.6 nm $\times$ 217.6 nm and divided into 128 $\times$ 128 voxels is considered [48]. Each voxel has dimensions of 1.7 nm $\times$ 1.7 nm, which corresponds to the typical size of the STZ. A uniaxial tensile test of the MG sheet is performed to verify the accuracy of the proposed model. The strain rate and strain increment are $1 \times 10^{-4}$ s$^{-1}$ and $1 \times 10^{-4}$, respectively. Periodic boundary conditions are applied to the four edges of the plate. The number of transformation modes and the activation energy are set to 20 and 5 eV, respectively.

Fig. B1 shows the stress-strain curves of MG sheets from the original mesoscale kMC model and our proposed model. We can observe that our results are in good agreement with those of the original mesoscale kMC model, while the small differences are due to the different random seeds in the kMC algorithm.

![Fig. B1. The stress-strain curves of MG sheets from the original mesoscale kMC model [48] and the proposed model.](image)

The original mesoscale kMC model, employing heterogeneous randomized catalogs and generation-dependent softening, successfully elucidates the intricate relationship between the shear band formation and the internal structural information. It underscores the pivotal role of softening in strain localization and shear band development. However, when applied to the study of NGs, the original mesoscale kMC model falls short of accurately representing the influence of grain size on the mechanical behavior of NGs, as observed in molecular dynamics simulations [28–32]. Despite our attempts to modify the model by assigning different values for transformation modes and activation energy barriers at the grain and interfaces in NGs (refer to Section 3.1 for details), this still indicates a need for a refined model to systematically investigate the influence of grain size on the mechanical behaviors of NGs.

Recognizing the reliance of the mesoscale model on detailed atomistic simulations, as emphasized by Zhao et al. [48], we are motivated to address microstructural changes in MG or NG during the shear band development process. Inspired by the aged-rejuvenation-glue-liquid (ARGL) model proposed by Shimizu et al. [61] which categorizes the shear band into four zones ranging from well-aged glass to near-liquid state, we envision a progressive change of local inelastic strain corresponding to these four zones. To operationalize this concept, in this study we introduce a variable characteristic strain and employ a sigmoid function to represent its evolution during the deformation process.

Intriguingly, molecular dynamics simulations (refer to Supplementary data for details) demonstrate that the magnitude of the characteristic strain
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